

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of:  
Kouichi KITAHATA et al.

Application No.: 10/571,582

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Art Unit: 1611

For: POROUS SILICA HAVING SUBSTANCE  
CARRIED THEREON

Examiner: ORWIG, Kevin S.

DECLARATION UNDER 37 CFR 1.132

COMMISSIONER FOR PATENTS  
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Sir:

I, Katsuyuki MIYAZAKI, residing in Mie-ken, Japan, hereby declares and states as follows:

1. That I am thoroughly familiar with the contents of U.S. Application Serial No. 10/571,582 filed on March 10, 2006, entitled POROUS SILICA HAVING SUBSTANCE CARRIED THEREON, its prosecution before the United States Patent and Trademark Office and the references cited therein.

2. I am a graduate of The Mie University Graduate School of Medicine, Department of Stem Cell and Developmental Biology and received a master's degree in the year 2009, majoring in developmental biology and technology.

3. That I have been employed in Taiyo Kagaku Co., Ltd. in the year 2009 and have been assigned to the Research Laboratories.

4. I have been involved in the research and development of mesoporous silica since 2009.

5. The following experiments were conducted by myself or under my direct supervision and control in order to compare and study the porous silica of the present invention with the lumpy mesoporous silica of WO 98/14399, in order to distinguish the present invention with the WO 98/14399.

## METHODS

### Synthesis of Porous Silica

#### Present Invention

A porous silica was prepared in accordance with the method of Preparation Example 1 of Porous Silica of the present invention.

Specifically, 50 g of No. 1 sodium silicate ( $\text{SiO}_2/\text{Na}_2\text{O} = 2.00$ ) manufactured by Nippon Chemical Industrial Co., LTD. was dispersed in 1000 ml of a 0.1 M aqueous solution of octadecyltrimethylammonium chloride [ $\text{C}_{18}\text{H}_{37}\text{N}(\text{CH}_3)_3\text{Cl}$ ], a surfactant, and the dispersion was heated at 70°C for 3 hours while stirring. Thereafter, while heating the mixture at 70°C and stirring, a 2 N hydrochloric acid was added to the dispersion to lower its pH to 8.5, and the mixture was further heated at 70°C for 3 hours while stirring. A solid product was temporarily filtered and re-dispersed in 1000 ml of ion-exchanged water while stirring. The procedures of filtration and dispersion-stirring were repeated

5 times, and thereafter the residue was dried at 40°C for 24 hours. The dried solid product was heated in nitrogen gas at 450°C for 3 hours, and thereafter the heated solid product was baked in air at 550°C for 6 hours to give a porous silica A.

#### WO 98/14399

A mesoporous silica was prepared in the same manner as described in Example 1-2 of WO 98/14399.

Specifically, 0.5 mol of anhydrous sodium metasilicate and 0.1 mol of stearyltrimethylammonium chloride were added to 1-L of ion-exchanged water to dissolve at 70°C. Further, 2N HCl was added to the solution, to adjust its pH to 8.5. Thereafter, rinsing with water was repeated 5 times, washed with acetone, and dried. This dry powder was baked at 700°C for 5 hours.

#### Evaluation Methods

Each of the porous silicas obtained above was subjected to electron micrographic observations and a test for confirming sustained release property of fragrance (change with time), to confirm the differences therebetween.

- 1) The difference in particle sizes of each of the porous silicas was confirmed by an electron microscope and particle size distribution analyzer (manufactured by BECKMAN-COULTER). The results are shown in FIG. 1 and Table I.
- 2) Using each of the porous silicas, a fragrance-holder of lavender oil was prepared. These holders were allowed to stand in a thermostat kept at 40°C for 1 to 3 weeks, and

the residual amount of the fragrance was measured with TG/DIA (thermoanalyzer) to confirm sustained release property in the long term. The results are shown in Table II.

3) In order to confirm the residual amount of the lavender oil of each of the fragrance holders using each of the porous silicas, sensory evaluation was conducted by 10 panelists, and evaluated in 5 ranks from 5 to 1, rank 5 being the strongest fragrance. An average thereof was obtained. The results are shown in Table III.

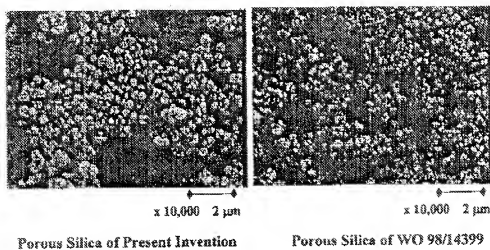


FIG. 1 Electron Micrographic Observations

**TABLE I: Average Particle Size of Each of Porous Silicas**

	Average Particle Size
Present Invention	400 nm
WO 98/14399	200 nm

**Table II: Evaluation of Sustained Release Property of Fragrance  
(Evaluated During 1 to 3 weeks)**

Residual Amount of Lavender Oil	1 week	2 weeks	3 weeks
Porous Silica of Present Invention	99.3%	98.9%	98.6%
Porous Silica of WO 98/14399 (Example 1-2)	81.3%	66.9%	59.2%

**Table III: Sensory Evaluation of Fragrance**

Sensory Evaluation of Lavender Oil	1 week	2 weeks	3 weeks
Porous Silica of Present Invention	3.8	3.2	3.3
Porous Silica of WO 98/14399 (Example 1-2)	4.8	3.7	3.3

It can be seen from the above evaluation results that the results of the residual amount of lavender oil of 99% after 1 to 3 weeks show the presence of fragrance upon sensory evaluation even after 3 weeks. Thus, the porous silica of the present application releases fragrance in a very small amount, i.e. shows excellent sustained release property.

DISCUSSION

In the evaluation method of sustained release property (weight change) of the lavender oil, a difference was clearly confirmed between the porous silica of the present invention and the porous silica of WO 98/14399. The differences in the property would be caused by the difference in the particle size. More exactly, the depth of the micropores would be larger in the porous silica of the present invention because of its larger particle size, leading to a larger depth of micropores.

**Statement Under 18 U.S.C. § 1001**

The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

Katsuyuki MIYAZAKI

Katsuyuki MIYAZAKI

January 11, 2011

Date